Good Vibrations: Terahertz Radiation Controls Material Properties

As reported in Nature, in a collaboration between the group of Robert Schoenlein at Berkeley Lab and researchers at Oxford University, ultrafast pulses of terahertz radiation were used to excite selected vibrations to change a manganite crystal from an electrical insulator into a conductor. This marks the first experimental demonstration that the excitation of a single vibrational mode can be used to induce phase changes in a crystal, and has important implications for the control of prized technological phenomena such as superconductivity and magnetoresistance.

Electrons are said to be "correlated" when the activity of one has an influence on its neighbors. In materials that are strongly correlated, even a subtle alteration of electronic activity can have an enormous impact on electrical and magnetic properties. For example, in the presence of a magnetic field, some strongly correlated materials will decrease electrical resistance by orders of magnitude, a phenomenon known as colossal magnetoresistance or CMR. Today's electronics industry is largely based on semiconductors that, under ordinary conditions, are weakly correlated. If the processing and control that is routinely achieved with these semiconductors could be achieved with strongly correlated materials, it could lead to a broad range of remarkable new technological possibilities.

To try to achieve this control on a fast time scale, many researchers have explored the dynamics of phase changes induced by ultrafast laser excitation to electronically excited states. But these studies have revealed little about the crucial ground state dynamics.

The Berkeley team set out to understand the phase change dynamics in the ground state of a CMR manganite ($Pr_{0.7}Ca_{0.3}MnO_3$), a manganese oxide with strongly correlated electrons. Below a critical temperature (~100 K) PCMO displays magnetoresistance, in which the crystal's electrical resistance can change 1,000 fold or more. To induce this change in resistance on an ultrafast time scale, the team exposed single crystals of manganite to femtosecond pulses of terahertz (10^{12} cps) radiation chosen to be resonant with a specific manganese-oxygen vibrational mode. To monitor changes in the conductance properties, the crystal was spectroscopically probed with femtosecond pulses of light that ranged in frequency between visible and infrared wavelengths – increased reflectivity of this second pulse signaled the transition from insulating to metallic.

The team found that a frequency of about 17 THz induced vibrations in the manganite crystal that resulted in a stretching of the electronic bonds that connect its manganese and oxygen atoms. This mild distortion of the crystal's geometry caused a profound change in its electronic properties; an increase in the crystal's electrical conductivity by five orders of magnitude on an ultrafast timescale.

This experiment shows that phase-changes in solid materials can be induced by selective vibrational excitation. Furthermore, it demonstrates that the dynamics of a phase change in a solid can be observed when the solid resides in the electronic ground state—the electronic state in which most chemical reactions and phase transitions take place.

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M. Rini, R. Tobey, N. Dean, J. Itatani, Y. Tomioka, Y. Tokura, R. W. Schoenlein, & A. Cavalleri "Control of the electronic phase of a manganite by mode-selective vibrational excitation" *Nature* **449**, 93 (2007).

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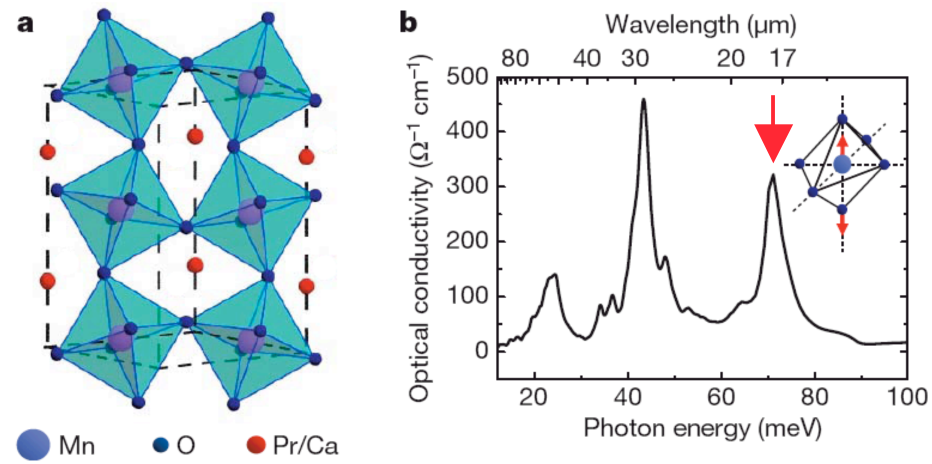
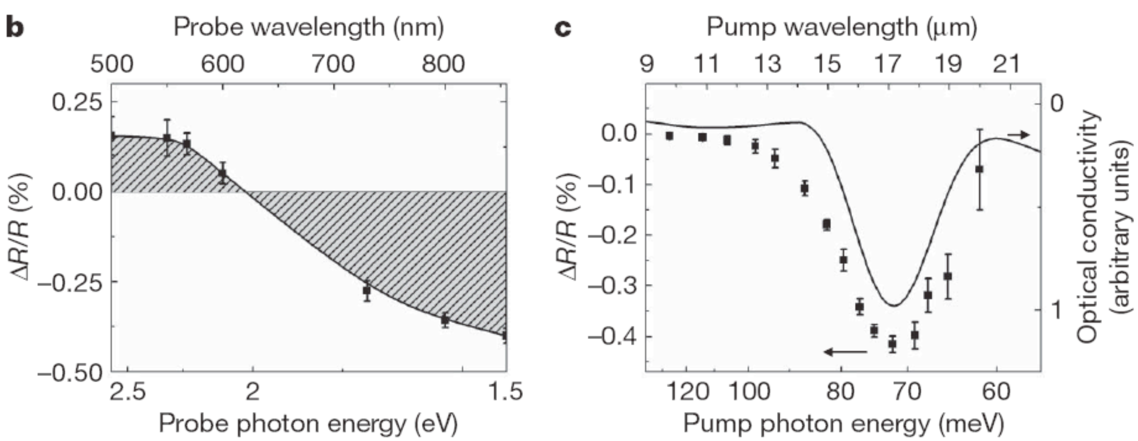
Colossal Magneto-Resistive (CMR) Manganite is a manganese oxide whose crystals have strongly correlated electrons. Below a critical temperature, ~100 K, CMR manganites display the phenomena known as magnetoresistance, in which the crystal's electrical resistance can change by 1,000x or more by applying a magnetic field.

Ultrafast Insulator-to-Metal Transition Initiated by Coherent Vibrational Excitation



Crystal structure *(left) and vibrational spectrum (right) of CMR manganite ($Pr_{0.7}Ca_{0.3}MnO_3$). The Mg-O vibration whose excitation caused the ultrafast resistance change is indicated by the arrow.*



Femtosecond pump–probe reflectivity

Left: Changes in reflectivity 1 ps after the THz excitation pulse. Shift of spectral weight towards longer wavelengths (shaded area) is a signature of the formation of the metallic phase.
Right: Reflectivity change at 1.5 eV, indicating metallic phase, as a function of pump wavelength (squares). Absorption spectrum of Mg-O vibration resonance at 17 THz (solid line). Both data sets peak at 17 THz, showing that selective excitation of this vibration is causing the resistance change.